The Piston Effect as a means to measure near-critical bulk viscosity

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Abstract :

Fluids close to their liquid-vapour critical point exhibit peculiar properties which have strong consequences on their hydrodynamics. Considerable experimental work has been conducted to measure the divergence of near-critical fluid's properties close to the critical point. But owing to their peculiar hydrodynamics, some of these quantities have proved difficult to access. One way out of this limitation has been the use of microgravity experiments, in which the effects of gravity could be suppressed. But even in the absence of gravity, near-critical fluids are subjected to very strong dynamical phenomena. In particular, it has been observed that the heat transfer in fluids near the liquid-vapour critical point is governed not only by diffusion, convection and radiation, but also by a thermo-mechanical coupling called the Piston Effect, discovered in the early 90s.

Among the least known properties close to the critical point is the bulk viscosity, which is expected to exhibit a very strong critical divergence. Despite this divergence, most existing theoretical models of the Piston Effect are based on non-viscous equations. Using equations recently developed for the hydrodynamics of viscous near-critical fluids, we propose a new indirect way of measuring bulk viscosity close to the critical point. This method is based on the use of a carefully monitored Piston Effect, acting as a probe and triggering a dynamic response in which the signature of bulk viscosity can be measured.

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1. INTRODUCTION

It is now a well-established fact that the heat transfer in the vicinity of the critical point is governed not only by diffusion, convection and radiation, but also by a thermo-mechanical coupling called the Piston Effect. This effect is responsible for the critical speeding up of the heat transfer, which contradicts the expectation of a critical slowing down of diffusive processes such as heat diffusion. The Piston Effect takes place when a confined near-critical fluid is submitted to a boundary heating : the heat diffuses in a thin boundary layer, which expands due to the very high compressibility of the fluid; this expansion drives in turn an isentropic compression of the bulk fluid, resulting in a homogeneous temperature rise. This effect takes place on a shorter and shorter time scale as the critical point is neared, so that a confined near-critical fluid behaves like a thermal short-circuit. Beyond the first contribution which pointed out this phenomenon (Onuki [1]), several teams have proposed more refined theoretical models, while space-borne experiments have given access to quantitative measurements of the dynamics of this effect (see [2] for a recent review).

In almost all the existing models, viscous effects have been systematically neglected. However, it is known that bulk viscosity diverges at the critical point (see Kadanoff and Swift [3], Kawasaki [4], Quentrec [5]), a divergence which has strong consequences on dynamical phenomena such as the Piston Effect. In a first attempt, Onuki [6] derived an explicit expression of the stress tensor of a near-critical fluid, and applied it to a few thermodynamic situations in which an influence of the bulk viscosity on the Piston Effect was predicted. But this contribution, based on global thermodynamic methods, relied (among other things) on the assumption of a completely homogeneous pressure in the fluid. Although this assumption is correct in the classical models of Piston Effect, it fails close to the critical point, as was first shown by Carlès [7]. A first hydrodynamic model of viscous Piston Effect was then proposed by Carlès [8], which took into account all the dynamic phenomena related to the existence of a diverging bulk viscosity close to the critical point.

This theoretical model predicted that close to the critical point, the bulk viscosity prevents the boundary layers from expanding, by creating a pressure gradient close to the heated walls which competes against viscous stresses. The result of this competition is a strong weakening of the Piston Effect, making the temperature relaxation by the Piston Effect slower and slower as the critical point is neared. Instead of the predicted Critical Speeding Up, one should observe a new regime of Critical Slowing Down close to the critical point.

In the present work, we first show how the influence of bulk viscosity on the Piston Effect can be summarized by the definition of a second typical time-scale, the viscous time-scale, which complements the now classical Piston Effect time-scale (as first defined by Onuki [1]). Relying on the existence of this second time-scale, we then propose a new experimental procedure which uses the viscous Piston Effect as a probe to measure near-critical bulk viscosity.

2. THE PROBLEM UNDER STUDY

As in [8], a closed cell is filled with a supercritical fluid, initially at rest and in thermal equilibrium, in a zero-g environment in order to suppress all convective motion. The temperature of one of the walls of the cell is increased, while the other walls are thermally insulated.

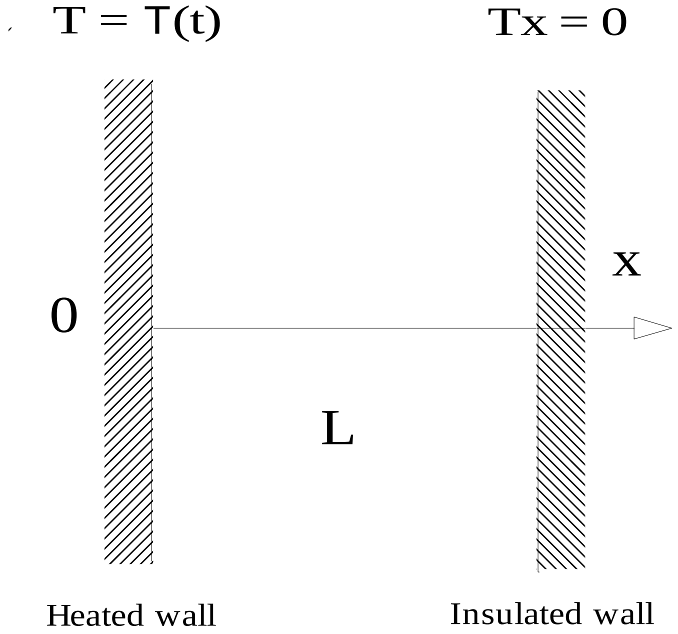


Figure 1: Problem under study

To conduct the calculation, the cell is assimilated to a mono-dimensional container between two infinite planar walls, separated by a distance L. Confined between the two walls is a near-critical fluid initially at rest, at critical density and at a temperature Ti slightly above the critical temperature Tc. At t = 0 (initial instant), a time-dependent temperature law T(t) is imposed at the left wall, while the right wall is thermally insulated (see figure 1). Using the model proposed in [8], we try to predict the time behavior of the density, the temperature, the pressure and the velocity of the fluid throughout the cell.

The equations which are used to model the fluid are Navier-Stokes equations, to which are added the equation of energy and an equation of state. Let us denote the critical coordinates by Tc, ρc and Pc. To represent the critical behavior of the fluid, the transport coefficients are written as the products of co-factors and powers of $\Delta T = (Ti - Tc) / Tc$:

$$Cv = Cv_0 \ \Delta T^{-\alpha}$$

$$\lambda = \lambda_0 \ \Delta T^{-x_\lambda}$$

$$\left(\frac{\partial \rho}{\partial P}\right)_T = \Gamma_0 \ \Delta T^{-\gamma}$$

$$\left(\frac{4}{3}\eta_S + \eta_B\right) \Big/ \rho_c = \theta_0 \ \Delta T^{-x_{\eta_B}}$$

In the above expressions, Cv, λ, η_S and η_B are respectively the heat capacity at constant volume, the thermal conductivity and the shear and bulk viscosity (written in the zero-frequency limit). α, x_λ, γ and $x_{\eta B}$ are the corresponding critical exponents (as an example, for a real fluid, $\alpha \approx 0.11$, $x_\lambda \approx 0.55$, $\gamma \approx 1.24$ and $x_{\eta B} \approx 1.92$). In what follows, quantities with the indice (0) will always refer to co-factors or products of co-factors.

The model in [8] is based on an asymptotic analysis of the equations, which are non-dimensionalized using the following parameters:

$$T^* = \frac{T}{T_c} \qquad P^* = \frac{P}{P_c} \qquad \rho^* = \frac{\rho}{\rho_c} \qquad u^* = \frac{u}{c} \qquad x = \frac{x}{L} \qquad t^* = \frac{t\,c}{L}$$

In the above expressions, c represents the celerity of sound, defined as :

$$c^2 = \frac{T_c}{\rho_c{}^2 Cv}\left(\frac{\partial P}{\partial T}\right)_\rho^2 + \left(\frac{\partial P}{\partial \rho}\right)_T \approx \frac{T_c}{\rho_c{}^2 Cv}\left(\frac{\partial P}{\partial T}\right)_\rho^2$$

(at the critical point, the isothermal compressibility becomes infinite).

The above definitions lead to the following equations (forgetting the stars):

$$\rho_t + (\rho u)_x = 0 \tag{1}$$

$$\rho u_t + \rho u u_x = -\, A\, \Delta T^{-\alpha} P_x + \varepsilon\, \Delta T^{-\alpha/2}\left(\eta_0\, \Delta T^{-x_{\eta B}} u_x\right)_x \tag{2}$$

$$\rho T_t + \rho u T_x = -\, B \Delta T^{\alpha} T u_x + \varepsilon\, \Delta T^{\alpha/2} \frac{\gamma_0}{Pr_0}\left(\Delta T^{-x_\lambda} T_x\right)_x + \varepsilon\, \Delta T^{3\alpha/2 - x_{\eta B}} B^2 \eta_0 u_x^2 \tag{3}$$

$$\delta P = C \delta T + D \delta \rho \tag{4}$$

(the equation of state being linearised). As one can see, the transport properties have been written as functions of $\Delta T = (Ti - Tc) / Tc$ alone. They are consequently regarded as constants once the initial conditions are given. Equations (1)-(4) are thus valid only if small disturbances from these initial conditions are considered, both in temperature and density.

In the above equations, the following non-dimensional parameters are defined :

$$A = \frac{P_c \rho_c}{T_c}\, Cv_0 \left(\frac{\partial T}{\partial P}\right)_\rho^2 \qquad B = \frac{1}{\rho_c Cv_0}\left(\frac{\partial P}{\partial T}\right)_\rho$$

$$C = \frac{T_c}{P_c}\left(\frac{\partial P}{\partial T}\right)_\rho \qquad D = \frac{\rho_c}{P_c\, \Gamma_0}$$

$$\varepsilon = \frac{\eta_{S0}}{\rho_c L\, C_0} \qquad \frac{\gamma_0}{Pr_0} = \frac{\lambda_0}{\eta_{S0}\, Cv_0} \qquad \eta_0 = \frac{\rho_c\, \theta_0}{\eta_{S0}}$$

η_{S0} and C_0 are respectively the co-factor for the shear viscosity and the sound velocity. γ_0 is the reference ratio of heat capacities (Cp_0 / Cv_0) and Pr_0 the reference Prandtl number based on co-factors. All these parameters are scaling factors which have no direct incidence on the final results of the calculation. Note that ABC = 1.

A, B, C, D, η_0 and γ_0/Pr_0 are order one parameters, while ε is much smaller than 1.

The initial conditions are then :

$$\begin{array}{ll} T(t=0) = 1 + \Delta T & \rho(t=0) = 1 \\ P(t=0) = P_i & u(t=0) = 0 \end{array} \qquad (5)$$

The boundary conditions are :

$$\begin{array}{ll} T(x=0,t) = \mathrm{T}(t) & \text{(non-dimensional temperature law at the left wall)} \\ u(x=0,t) = 0 & \text{(contact between the fluid and the left wall)} \\ T_x(x=1,t) = 0 & \text{(adiabatic right wall)} \\ u(x=1,t) = 0 & \text{(contact between the fluid and the right wall)} \end{array} \qquad (6)$$

3. ASYMPTOTIC ANALYSIS

The above system has already been solved by Carlès [8] in the case of an imposed constant heat flux at the left wall. The resolution method was based on asymptotic expansions with the limits $\varepsilon \to 0$ and $\Delta T \to 0$. It was shown that under these asymptotic conditions, the heat diffusion term in the energy equation is negligible everywhere except close to the heated wall. The system is thus asymptotically singular in space, at x = 0. In order to get rid of this singularity, a boundary layer was defined using a new space variable $z = x / \delta$ where $\delta(\varepsilon, \Delta T)$ is the boundary layer thickness (much smaller than 1). The equations in the boundary layer where matched with the equations far from the wall (the bulk) using the method of Matched Asymptotic Expansions. The solution was sought on the typical time-scale of temperature relaxation, which is defined by the typical time-scale on which temperature elevation is of the same order in the boundary layer and in the rest of the fluid (the bulk). This led to the definition of the time variable $\tau = e\, t$ where $e = e(\varepsilon, \Delta T)$ is the inverse of the temperature relaxation characteristic time, in its non-dimensional form. Finally, the existence of two independent asymptotic parameters ε and ΔT led to the presence of a singularity in the space of parameters (i.e. the solution is dependent on the order chosen in the two limits $\varepsilon \to 0$ and $\Delta T \to 0$). It was shown that this singularity could be resolved and a solution found for an arbitrary pair (ε, ΔT) provided the following condition was fulfilled:

$$\varsigma = \frac{\Delta T}{\varepsilon^{2/\chi}} = O(1)$$

with $\chi = 2\gamma + x_\lambda + x_{\eta B} - 2\alpha$.

The same method is applied here, with the new boundary conditions (6). The details of the calculations can be found in [9], and will not be repeated here. The obtained solution can be described as follows:

When the boundary temperature is increased, a thin thermal boundary layer forms in the fluid close to the wall, thanks to heat diffusion. This thin boundary layer expands due to the large compressibility of the fluid, thus driving a macroscopic velocity in the fluid cell. Due to this velocity, viscous stresses appear in the boundary layer which compete against its free expansion. As a result, a pressure gradient starts to build up in the boundary layer. For parameters such that $\zeta >> 1$ (which means not too close to the critical point), the viscous constraints are negligible: the classical Piston Effect is found. For parameters such that $\zeta << 1$ on the contrary, the viscous stresses strongly affect the expansion of the boundary layer, and the Piston Effect enters a regime of Critical Slowing Down.

The mathematical details of this solution will not be given here, but will be detailed in a forthcoming publication. We only present here a prominent result, which is the relationship between the imposed boundary temperature $\mathsf{T}(t)$ and the bulk mean temperature $T_B(t)$. This relationship is obtained under the form of the following Laplace transforms (s being the Laplace complex parameter):

$$T_B(s) = \mathsf{T}(s) \frac{1}{1 + \sqrt{\dfrac{D\, Pr_0\, L}{B\, C\, \gamma_0\, c\, \varepsilon}\, \Delta T^{\gamma + x_\lambda - \frac{3}{2}\alpha}\, s + \dfrac{\eta_0\, Pr_0\, L^2}{\gamma_0\, c^2}\, \Delta T^{x_\lambda - \alpha - x_{\eta B}}\, s^2}}$$

Using the definition of the various non-dimensional parameters, the above solution can be rewritten as:

$$T_B(s) = \mathsf{T}(s) \frac{1}{1 + \sqrt{\dfrac{s}{\Omega_c} + \left(\dfrac{s}{\Omega_v}\right)^2}} \tag{7}$$

where Ω_c and Ω_v are two typical frequencies, defined as:

$$\Omega_c = -\frac{Tc\, \lambda}{\rho_c^{\ 3}\, L^2\, Cv^2} \left(\frac{\partial \rho}{\partial T}\right)_P \left(\frac{\partial P}{\partial T}\right)_\rho \tag{8}$$

$$\Omega_v^{\ 2} = \frac{Tc\, \lambda}{\rho_c^{\ 2}\, L^2\, Cv^2 \left(\eta_B + \dfrac{4}{3}\eta_s\right)} \left(\frac{\partial P}{\partial T}\right)_\rho^2 \tag{9}$$

4. DISCUSSION OF THE RESULTS

A close examination at the above frequencies reveals that they are the inverse of two typical times which can be expressed in a simple way:

$$t_{PE} = \frac{1}{\Omega_c} = \frac{t_D}{(\gamma - 1)^2} \tag{10}$$

$$t_v = \frac{1}{\Omega_v} = \sqrt{t_{PE} \frac{\eta_B}{\kappa_T}} \tag{11}$$

where κ_T is the thermal diffusivity of the fluid. t_{PE} can be recognized as the classical Piston Effect time-scale, as first identified by Onuki [1]. We thus prove that, contrarily to what was assumed so far, the definition of t_{PE} is not sufficient to completely characterize the Piston Effect: a second time-scale t_v is also necessary. This later time-scale represents the typical relaxation time imposed by the bulk viscosity. It affects the global heat transfer in a drastic way close to the critical point, as first shown in [7]. This influence can be observed if one plots t_{PE} and t_v as functions of the reduced temperature, as has been done on figure 2 in the case of CO_2 in a 10 mm container.

It can be observed that, while t_{PE} goes to zero when the critical point is neared, t_v goes to infinity. As is clear from equation (7), the temperature relaxation follows the longest of these two time-scales. Hence, after a first regime of critical speeding up, temperature relaxation undergoes a regime of critical slowing down close to the critical point.

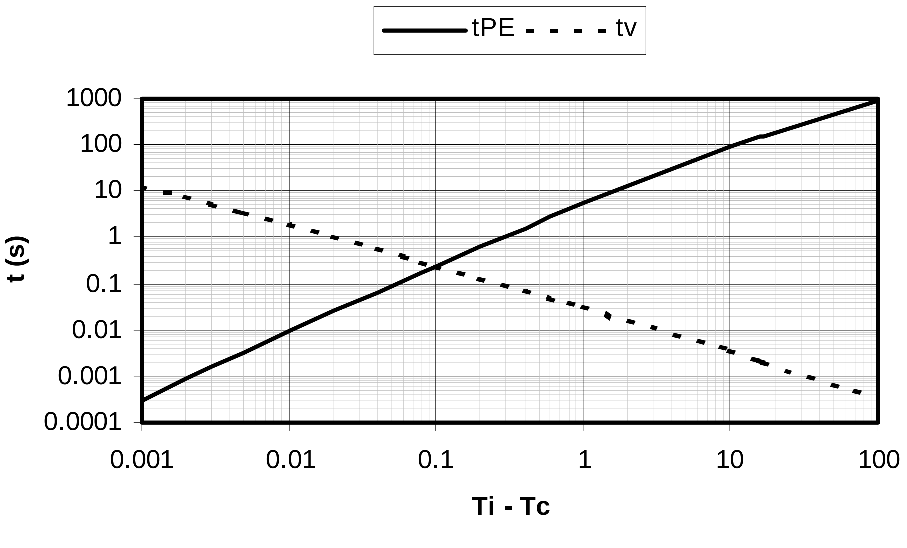


Figure 2: t_{PE} and t_v as functions of Ti – Tc, for CO_2 in a 10 mm cell

In the so-called “classical” regime of the Piston Effect, characterized by time-scale t_{PE}, $\Omega_c << \Omega_v$, so that a first approximation of the bulk temperature law is:

$$T_B(s) \approx T(s)\,\frac{1}{1+\sqrt{\dfrac{s}{\Omega_c}}} \qquad (12)$$

which can be inverted in :

$$T_B(t) \approx T(t) * \frac{1}{t_{PE}}\left[\sqrt{\frac{1}{\pi\,(t/t_{PE})}} - e^{(t/t_{PE})}\mathrm{erfc}\left(\sqrt{t/t_{PE}}\right)\right] \qquad (13)$$

where * stands for the convolution product. It can easily be seen that equ.(13) leads to the now classical solution for the non-viscous Piston Effect (see for instance [7] and [10]). The non-viscous Piston Effect is obtained as a particular case of the present theory, far from the critical point where bulk viscosity is negligible.

In the so-called "viscous" regime of the Piston Effect, one has on the contrary $\Omega_c >> \Omega_v$, so that a first approximation of the bulk temperature law is:

$$T_B(s) = T(s)\,\frac{1}{1+\dfrac{s}{\Omega_v}} \qquad (14)$$

which can be inverted in:

$$T_B(t) \approx T(t) * \frac{1}{t_v}\,e^{-(t/t_v)} \qquad (15)$$

Now, the temperature relaxation is no longer governed by t_{PE} but by t_v. One can easily recognize in the above relations (15) the classical response of a system subjected to a first-order low-pass filter: the bulk temperature is simply equal to the boundary temperature filtered through a first-order low-pass filter of cut-off frequency Ω_v. As Ω_v goes to zero at the critical point, the temperature relaxation undergoes a critical slowing down in the viscous regime.

5. A METHOD FOR MEASURING NEAR-CRITICAL BULK VISCOSITY

The filtering property described above can be used in order to measure the near-critical bulk viscosity close to the critical point. The idea is to use the Piston Effect as a carefully controled perturbation, the dynamic response of which will be measured to deduce the value of transport properties. It is thus based on the principle of inverse methods.

The experimental setup should be one of a fluid cell whose walls are thermostated, filled with a near-critical fluid at critical density and placed in microgravity conditions. The typical size of the cell should be chosen so as to insure that the fluid is in the viscous regime of Piston Effect for the whole range of relevant reduced temperatures (Ω_c/Ω_v is proportional to L, so that by selecting a proper cell dimension, one can decide at what reduced temperature the Piston Effect in the cell will enter the viscous regime). In this viscous regime, the transfer function between the boundary temperature and the bulk temperature is one of a first-order low-pass filter of cut-off frequency Ω_v. Ω_v is a direct function of the bulk viscosity (see equ.(9)), so that bulk viscosity can be deduced from an accurate measurement of the cut-off frequency. In order to do so, an AC heating of frequency ω should be imposed on one of the walls of the cell, while the temperature of the heated wall $\mathsf{T}(t)$ and the bulk temperature $T_B(t)$ are measured as functions of time. By sweeping ω over a large range of frequencies, the cut-off frequency Ω_v could be found by analyzing both the ratio of the amplitude of the oscillating temperatures $\mathsf{T}(t)$ and $T_B(t)$, and their phase-lag. Indeed, if $\mathsf{T}(t)$ is of the following form:

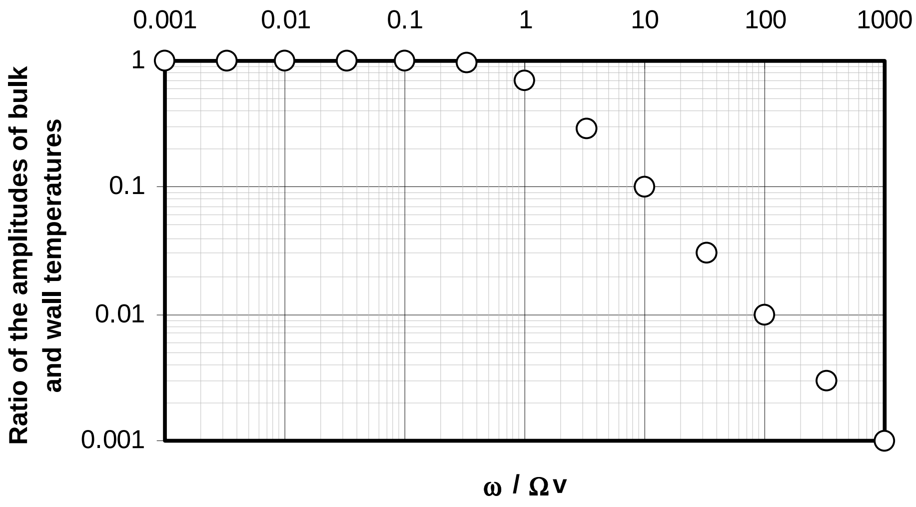
$$\mathsf{T}(t) = \Theta \sin(\omega t)$$

Then according to (14), $T_B(t)$ will be:

$$T_B(t) = \Theta \frac{\Omega_v}{\sqrt{\Omega_v^2 + \omega^2}} \sin(\omega t + \varphi)$$

with $\varphi = - \tan^{-1} (\omega/\Omega_v)$

On figure 3 can be seen the evolution with frequency of the amplitude ratio and phase-lag between $\mathsf{T}(t)$ and $T_B(t)$.



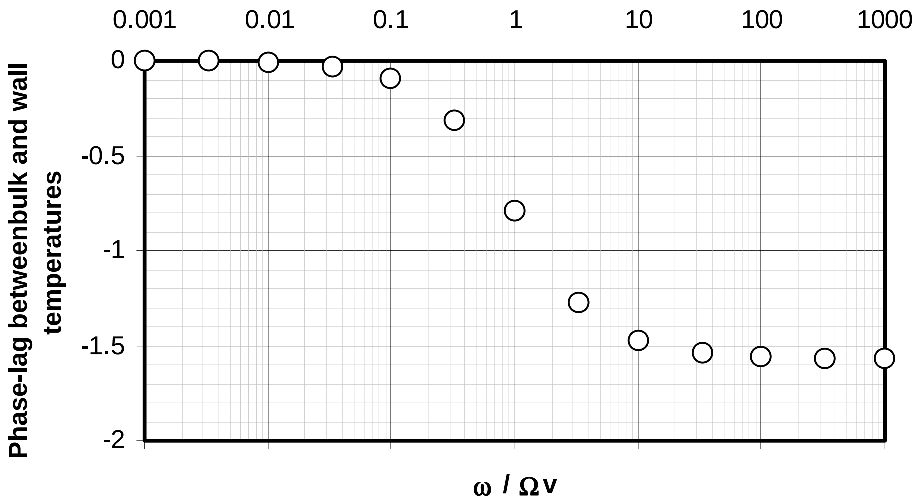


Figure 3: Ratio of amplitudes and phase-lag between T(t) and T_B(t)

As can be observed on figure 3, the amplitude drop at the cut-off frequency associated to the phase-lag should permit a precise determination of the value of Ω_v, and consequently of η_B, the bulk viscosity.

6. CONCLUSION

In the present work, we have shown that the Piston Effect is not governed by one but by two typical time-scales, one of which is the classical Piston Effect time-scale, while the other measures the time response of the boundary layer expansion when submitted to viscous constraints. By studying the temperature dependence of these two time-scales, it has been shown that the first regime of critical speeding up of the Piston Effect should be followed by a regime of critical slowing down, when viscous constraints start preventing the free expansion of the boundary layers. A first viscous model of Piston Effect by Carlès [8] has been extended to an arbitrary temperature law. This extension has shown that close to the critical point, in the viscous regime of critical slowing down, the bulk temperature follows the boundary temperature after being subjected to a low-pass first-order filter, the cut-off frequency of which has been predicted as a function of bulk viscosity. This later property could be used as an indirect way of measuring the bulk viscosity close to the critical point. The Piston Effect in this case would not be considered as a thermomechanical perturbation of the experimental conditions, but on the contrary as a probe used to measure physical quantities.

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